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Reflectance of nonlocal conducting superlattices

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We calculate the p-polarized reflectance spectra of a semi-infinite superlattice made up of alternating spatially dispersive conducting layers, accounting exactly within a hydrodynamic model, for the propagation of plasma waves. Besides the expected structure originated from the infinite-superlattice bulk-plasma resonances, we find an extra series of peaks which arise from the semi-infinite-superlattice surface resonances. The latter originate from standing plasma waves at the surface layer, whose resonant frequencies are larger than those in the interior layers due to confinement effects.

I. INTRODUCTION

In a previous paper,[1] a formalism was developed to calculate the dispersion relations of the p-polarized electromagnetic normal modes of a superlattice made up of nonlocal conducting layers described by hydrodynamic models of electron dynamics. This formalism accounts for the coupling to longitudinal waves by employing an enlarged 4×4 transfer matrix instead of the usual 2×2 matrix commonly employed in multifilm optics,[2] and leads to a general analytic expression useful for different choices of the so-called additional boundary conditions (ABC's).[3–6] This formalism was first applied to infinite periodic superlattices.[1] Their normal modes display a very rich spectra due to their manifold possible compositions. For example, there are SP-SP modes made up of surface plasmons (SP) propagating along each interface but intercoupled through their evanscent fields. There are also GP-ET modes due mainly to guided plasmons (GP) in the low-density layers coupled among themselves by the evanescent transverse (ET) waves they induce in the high-density layers. Furthermore, for each frequency there are two kinds of modes; one is mostly transverse and the other mostly longitudinal. Thus, there are twice as many modes than found by other workers.[7,5] The formalism above has recently been applied to quasiperiodic superlattices[8] where it led to critical modes with a Cantor-like spectra.

The multitude of modes mentioned above might be probed using a spectroscopy such as optical reflectance or electron-energy loss. The purpose of this paper is to extend the 4×4 transfer-matrix formalism to calculate the p-polarized reflectance spectra of the semi-infinite superlattice. There are previous reflectance calculations for conducting superlattices. Some assumed the conducting media were local.[9] Later, superlattices of alternating local insulators and spatially dispersive layers were considered, not only for conducting,[10,11] but also for excitonic semiconductor layers.[12] The reflectance of conductor-conductor heterostructures has been calculated for frequencies below the plasma frequency of the more dense layers by neglecting the spatial dispersion of the latter[13] since plasma waves would not propagate within them, and, unlike transverse waves, they would decay sharply. The nonlocality of both conducting layers was taken into account in Ref. 14, where unfortunately, the approach to the reflectance calculation is flawed.[15]

In the present paper we calculate the surface impedance and the reflectance of a semi-infinite superlattice by superposing both kinds of bulk modes in such a way as to obey at the superlattice-vacuum interface the additional boundary condition brought about[6] by the interior conductor-conductor ABC's. The theory is developed in Sec. II, some results are presented in Sec. III, and Sec. IV is devoted to conclusions.

II. THEORY

We consider a semi-infinite superlattice made up by stacking along the Z direction alternating layers of conductors a and b characterized within the hydrodynamic model by their widths d_i, plasma frequencies ω_i, stiffness constants β_i, relaxation times τ_i, and bound-electron dielectric functions $\epsilon_i^B(\omega)$, where i takes the values a and b. Their transverse (T) and longitudinal (L) frequency ω and wave-vector $\mathbf{q}$ dependent dielectric functions are taken to be

$$\epsilon_i^T(\omega) = \epsilon_i^B(\omega) - \frac{\omega_i^2}{\omega^2+i\omega/\tau_i}, \quad (1)$$

$$\epsilon_i^L(\omega,\mathbf{q}) = \epsilon_i^B(\omega) - \frac{\omega_i^2}{\omega^2+i\omega/\tau_i-\beta_i^2 q^2}. \quad (2)$$

For p-polarized light propagating along the X-Z plane we define, as in Ref. 1, a 4×4 transfer matrix M so that

$$\begin{pmatrix} E_x \\ B_y \\ \mu_i j_z \\ 4\pi i\nu_i\rho \end{pmatrix}_{z+d} = M \begin{pmatrix} E_x \\ B_y \\ \mu_i j_z \\ 4\pi i\nu_i\rho \end{pmatrix}_z , \quad (3)$$

where $d = d_a + d_b$ is the superlattice period, $\mathbf{E}$ is the electric field, $\mathbf{B}$ is the magnetic field, $\mathbf{j}$ is the conduction-electrons' current density, and ρ is their density deviation from equilibrium. The parameters μ_i and ν_i are obtained from the ABC's in such a way as to render the column vector $(E_x, B_y, \mu_i j_z, 4\pi i\nu_i\rho)^T$ continuous at all interior interfaces. The transfer matrix can be simply calculated as the product $M = M_b M_a$ of the transfer matrices of each layer, for which expressions were given in Eqs. (13)–(15) of Ref. 1.

The normal modes of the corresponding infinite superlattice are given by the eigenvectors of M and their dispersion relation is obtained by equating the corresponding eigenvalue to e^{ipd}, where p is Bloch's wave vector. The result is a quadratic equation in $\cos(pd)$ [Eqs. (20)–(23) of Ref. 1] and therefore has two pairs $\pm p$ of solutions for each frequency ω and wave-vector projection $\mathbf{Q} = (Q,0,0)$ unto the superlattice interfaces.

A wave incident on the surface can excite two outgoing modes[15] within the semi-infinite superlattice, besides a reflected wave. The transmitted waves are those that have $Q = \cos(\theta)\omega/c$ and $\mathrm{Im}(p) > 0$, where θ is the angle of incidence. Therefore, the fields within the superlattice and near its surface can be written as

$$\begin{pmatrix} E_x \\ B_y \\ \mu_a j_z \\ 4\pi i\nu_a\rho \end{pmatrix}_{z=0} = c_1 V_1 + c_2 V_2, \quad (4)$$

where $V_1 = (E_x, B_y, \mu_a j_z, 4\pi i\nu_a\rho)_1^T$ and $V_2 = (E_x, B_y, \mu_a j_z, 4\pi i\nu_a\rho)_2^T$ are the two outgoing eigenvectors of M, c_1 and c_2 are their as yet undetermined amplitudes, and we assumed that the first conductor is of type a and the second of type b (otherwise we would simply change M to $M_a M_b$). Three boundary conditions are now required to solve for the amplitude of the reflected wave. As usual, two of them are the continuity of the parallel components of the electric and magnetic fields E_x and B_y. The remaining boundary condition is determined[6] by the ABC's imposed at the conductor-conductor interfaces.

For definiteness, we choose $\mu_i = 1$ and $\nu_i = \beta_i^2/\omega_i^2$, which correspond to the continuity of the normal components of the conduction-electron current density and of the energy flux.[3] In this case, the surface[6] ABC is given by $j_z(0) = 0$, and Eq. (4) leads to

$$c_1/c_2 = -j_z^{(2)}/j_z^{(1)}. \quad (5)$$

The surface impedance $Z_p = E_x(0)/B_y(0)$ is then given by

$$Z_p = \frac{j_z^{(2)}E_x^{(1)} - j_z^{(1)}E_x^{(2)}}{j_z^{(2)}B_y^{(1)} - j_z^{(1)}B_y^{(2)}}, \quad (6)$$

from which the optical properties follow as usual. For instance, the reflection amplitude is $r_p = (\cos\theta - Z_p)/(\cos\theta + Z_p)$ and the reflectance is $R_p = |r_p|^2$. Equations (5) and (6) are also valid for the ABC's of Ref. 5; the results for those of Ref. 4 are obtained from Eqs. (5) and (6) by replacing j_z by ρ. The approach presented above has been shown to be equivalent to that of Ref. 14 in the bulk of the superlattice,[15] and to amend it at the surface of the semi-infinite system.

III. RESULTS

In Fig. 1 we show the reflectance of a semi-infinite superlattice on which p-polarized light is incident at an angle of $\theta = 70°$. We chose $\omega_a = \sqrt{2}\omega_b$, that is, we take the surface layer to be twice as dense as the second one. The widths are taken as $d_a = d_b = 0.25c/\omega_b$, the Fermi velocity is $v_b^F = 0.01c$, and we include a small dissipation $\omega_b\tau = 1000$. We denote this system as ab. In this case, the reflectance is in close correspondence to the dispersion relations of the superlattice. Figures 2 and 3 exhibit these dispersion relations for the modes of mostly transverse (MT) and longitudinal (ML) character, respectively. The classification was done according to the relative values of the transverse and longitudinal contributions to the energy flux[3] $S_z^T = (c/8\pi)\mathrm{Re}(E_x B_y^*)$ and $S_z^L = (2\pi\beta_a^2/\omega_a^2)\mathrm{Re}(j_z\rho^*)$. The mostly transverse and longitudinal modes are usually those with the largest and the smallest decay lengths, respectively.

The reflectance spectra display a series of sharp oscillations superimposed on two broad minima (Fig. 1). These broad minima, present also in the local calculation, correspond to excitation of two coupled-surface-plasmon bands[13] which can be seen in Fig. 2. The surface-plasmon bands are broken into several bands due to coupling with bulk-plasma resonances confined within the low-density b

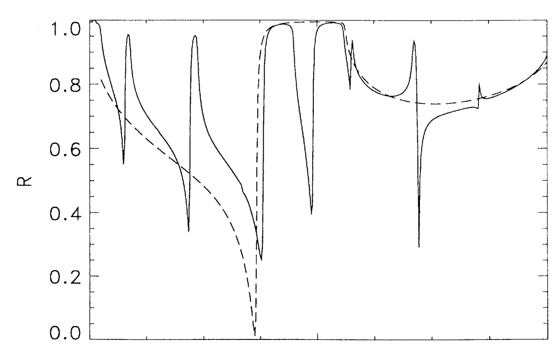

FIG. 1. Nonlocal (solid) and local (dashed) reflectance of a semi-infinite conducting superlattice-vacuum interface in the frequency region between the plasma frequencies of the two components. The surface layer has the larger of the two densities. The parameters are given in the text.

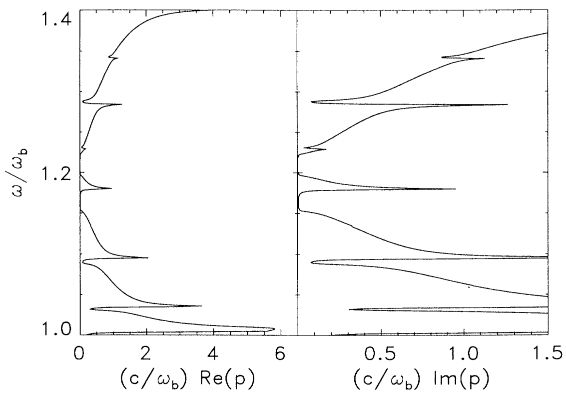

FIG. 2. Dispersion relation of the mostly transverse MT normal modes that can couple to light incident from vacuum at an angle $\theta = 70°$, for the same superlattice as in Fig. 1.

layers.[1] The resonance frequencies are close to the single film quantization condition $\omega_n^2 = \omega_b^2 + \beta_b^2[Q^2 + (n\pi/d_b)^2]$ with n an odd integer. These results are similar to those of Ref. 13 where the spatial dispersion of the high-density layers was neglected. However, in the present case the resonances are appreciably redshifted due to the finite decay length δ_a of plasma waves in the a layers. The resulting relaxation in the boundary conditions is equivalent to an increase in the width of the b layers by a quantity of the order of δ_a.[13]

The line shape shown in the right side of Fig. 1 has regions where the reflectance changes gradually as ω grows, followed by sharp jumps. A similar behavior is found in the left side of Fig. 1 as ω diminishes. This behavior can be understood in terms of the dispersion relation of the MT modes of Fig. 2. The reflectance is large whenever there is no propagating mode ($p' \approx 0$ and p'' large) and also when there is a propagating mode but with a large surface impedance mismatch ($p'' \approx 0$ but p' large). If p' diminishes gradually, the reflectance follows suit, and when p' approaches and attains 0 there is a deep minimum followed by a sharp increase. The ML modes have very flat bands at energies for which the MT modes also exhibit structure, so they do not generate extra features.

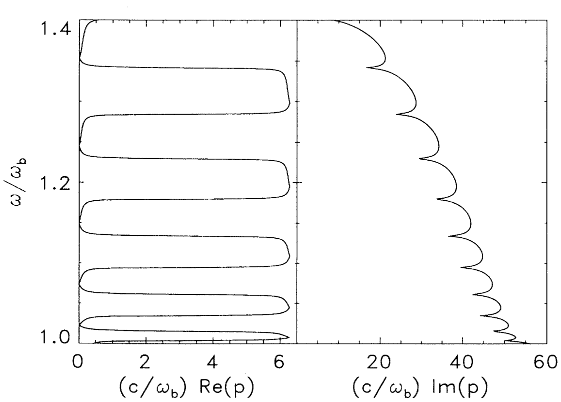

FIG. 3. Dispersion relation of the mostly longitudinal ML normal modes that can couple to light incident from vacuum at an angle $\theta = 70°$, for the same superlattice as in Fig. 1.

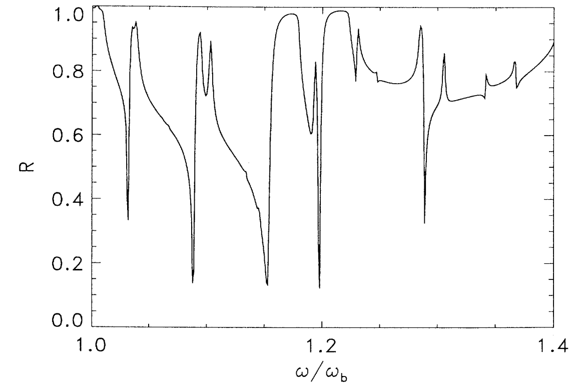

FIG. 4. Reflectance of a semi-infinite nonlocal conducting superlattice. The parameters are the same as in Fig. 1, except that the first layer has the lower of the two densities.

Figure 4 displays the reflectance spectra of a similar superlattice as that in Fig. 1, but for which the a and b layers have been permuted; that is, the surface layer has a lower density than the second layer. We denote this system as ba to distinguish it from the previous superlattice ab. Strikingly, the spectra exhibit twice as many peaks as that of Fig. 1. The position of every second minimum coincides with a minimum of Fig. 1, but is accompanied by a new structure at a higher frequency. To understand the origin of these extra features, in Fig. 5 we plotted the induced charge-density profile $|\rho(z)|$ for selected frequencies and for both the ab and the ba superlattices. We remark that the charge density corresponding to the new peaks (solid line in the lower panel of Fig. 5) has a very large amplitude in the first layer, revealing a surface resonance, in contrast to the behavior of the previous structures. The existence of these surface resonances may be explained as follows. Since the surface layer is

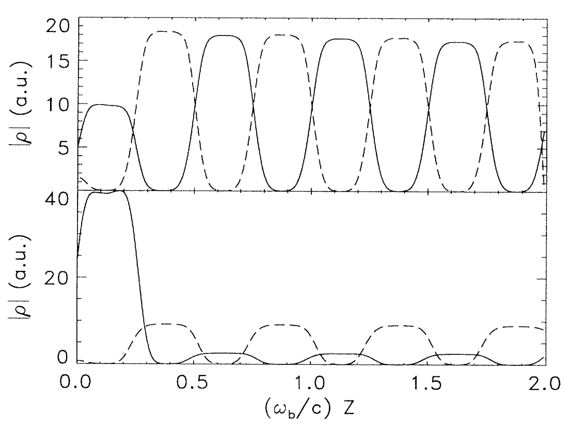

FIG. 5. Envelope of the magnitude of the induced charge density profile $|\rho(z)|$ for the same superlattices as in Figs. 1 (dashed) and 4 (solid), for a frequency $\omega = 1.086\,\omega_b$ near a reflectance minimum in both Figs. 1 and 4 where coupling to a propagating mode occurs (upper panel), and for $\omega = 1.103\,\omega_b$ which corresponds to a maximum in Fig. 4 only (lower panel), where a surface resonance is excited. To avoid cumbersome details, we do not display the oscillations of $\rho(z)$ within each layer.

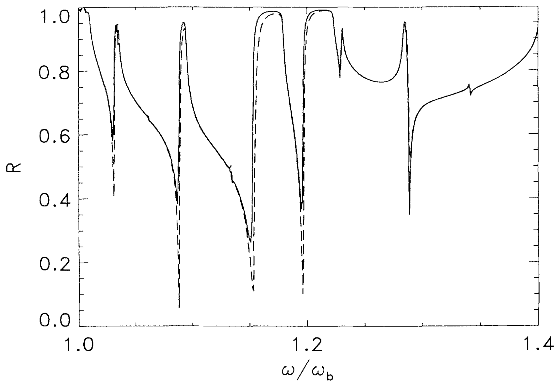

FIG. 6. Approximate reflectance of a semi-infinite nonlocal superlattice for the case *ab* in which the first layer is more dense (solid) and for the case *ba* in which the first layer is less dense (dashed). In the calculation the ML mode was neglected.

bounded by vacuum on one side, its plasmons can only spill over into the higher-density conductor on its other side, where the plasmon-decay length is finite. Therefore, the first-layer's effective width is smaller than that of the inner layers, which may spill over both sides. Consequently, the surface layer resonates at frequencies higher than the inner-layer resonances, but smaller than those of an isolated film. In contrast, in system *ab* all of the lower-density layers are bounded by similar conductors on either sides, and resonate at similar frequencies.

To clarify the relative contributions of both kinds of modes MT and ML to the reflectance spectra, we have performed calculations for the same systems as above, but purposefully neglecting the presence of the ML modes. In this case, ABC's cannot be satisfied at the surface, and the surface impedance is no longer given by Eq. (6), but simply by

$$Z_p = \frac{E_x^{(1)}}{B_y^{(1)}}, \tag{7}$$

where we assumed that the eigenvector 1 corresponds to the MT mode. In Fig. 6 we show the reflectance obtained within this approximation. Notice that for system *ab* the approximate calculation agrees closely with Fig. 1, whereas in case *ba* the additional structure of Fig. 4 is not reproduced. As expected, the violation of the ABC at the surface due to the neglect of the ML mode is more important when plasmons propagate within the surface layer than when they do not.

IV. CONCLUSIONS

We have developed a procedure for the calculation of the *p*-polarized reflectance spectra of semi-infinite nonlocal conducting superlattices by extending a previous 4×4 transfer-matrix formalism,[1] incorporating the manifold electromagnetic modes sustained by the superlattice. We performed calculations for a model superlattice confining our attention to frequencies between the plasma frequencies of both components. Besides the broad structures due to excitation of coupled surface-plasmon bands, a series of resonances was obtained at frequencies related to those of standing plasma waves circumscribed within the lower-density structures. These resonances are redshifted from those found in an isolated conducting film and in a local-nonlocal superlattice due to the spillover of the plasma-wave amplitude into nearby layers. Since the confinement of the plasma-waves is stronger in the surface layer, an additional series of surface resonances is obtained when the first layer has the lower density. A simpler calculational procedure that neglects the presence of the mostly longitudinal superlattice modes reproduces many of the features of the spectra, although it fails to produce the surface resonances.

ACKNOWLEDGMENTS

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[1]W. Luis Mochán and Marcelo del Castillo-Mussot, Phys. Rev. B **37**, 6763 (1988).
[2]E. Hecht and A. Zajac, *Optics* (Addison Wesley, Reading, MA, 1974), Sect. 9.9.
[3]F. Forstmann and H. Stenschke, Phys. Rev. Lett. **38**, 1365 (1977); Phys. Rev. B **17**, 1489 (1978).
[4]A. D. Boardman and R. Ruppin, Surf. Sci. **112**, 153 (1981).
[5]G. L. Eliasson, G. F. Giuliani, and J. J. Quinn, Phys. Rev. B **31**, 6534 (1985).
[6]M. del Castillo-Mussot and W. Luis Mochán, Solid State Commun. **62**, 555 (1987).
[7]M. Babiker, J. Phys. C **19**, L773 (1986).
[8]Wei-guo Feng, Nia-hua Liu, and Xiang Wu, Phys. Rev. B **43**, 6893 (1991).
[9]R. E. Camley and D. L. Mills, Phys. Rev. B **29**, 1695 (1984); G. F. Giuliani, J. J. Quinn, and R. F. Wallis, J. Phys. (Paris) Colloq. **45**, C5-285 (1984); R. Szenics, R. F. Wallis, G. F. Giuliani, and J. J. Quinn, Surf. Sci. **166**, 45 (1986).
[10]W. L. Mochán, M. del Castillo-Mussot, and R. G. Barrera, Phys. Rev. B **35**, 1088 (1987).
[11]P. Apell and O. Hunderi, in *Handbook of Optical Properties of Solids II*, edited by E. D. Palik (Academic, San Diego, 1991), p. 97.
[12]G. H. Cocoletzi and W. Luis Mochán, Phys. Rev. B **39**, 8403 (1989); G. H. Cocoletzi, A. Ramírez Perucho, and W. Luis Mochán, *ibid.* **44**, 11 514 (1991).
[13]M. del Castillo and W. L. Mochán, Phys. Rev. B **36**, 1779 (1987).
[14]M. Abraham, U. Trutschel, and A. Tadjeddinne, Opt. Commun. **28**, 369 (1986); U. Trutschel, M. Golz, and M. Abraham, Phys. Status Solidi B **151**, 383 (1989).
[15]W. L. Mochán, M. del Castillo-Mussot, and R. A. Vázquez-Nava, Phys. Status Solidi B (to be published).